

Isolation of proximity-induced triplet pairing channel in a superconductor/ferromagnet spin valve

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(Dated: March 31, 2016)

We have studied the proximity-induced superconducting triplet pairing in $\text{CoO}_x/\text{Py1}/\text{Cu}/\text{Py2}/\text{Cu}/\text{Pb}$ spin-valve structure (where $\text{Py} = \text{Ni}_{0.81}\text{Fe}_{0.19}$). By optimizing the parameters of this structure we found a triplet-channel assisted full switching between the normal and superconducting states. To observe an “isolated” triplet spin-valve effect we exploited the oscillatory feature of the magnitude of the ordinary spin-valve effect ΔT_c in the dependence of the Py2-layer thickness d_{Py2} . We determined the value of d_{Py2} at which ΔT_c caused by the ordinary spin-valve effect (the difference in the superconducting transition temperature T_c between the antiparallel and parallel mutual orientation of magnetizations of the Py1 and Py2 layers) is suppressed. For such a sample a “pure” triplet spin-valve effect which causes the minimum in T_c at the orthogonal configuration of magnetizations has been observed.

The superconducting spin-valve effect consists of different degree of suppression of superconductivity in the F1/F2/S or F1/S/F2 thin film multilayer constructions at parallel (P) and antiparallel (AP) mutual orientation of magnetizations of the F1 and F2 ferromagnetic layers. The superconducting spin valves based on the superconductor/ferromagnet (S/F) proximity effect offer a playground to explore fundamental aspects of interplay between superconductivity and magnetism and also promise applications as passive devices of the superconducting spintronics. The latter construction should be operational upon application of a small external magnetic field. Many experimental works were performed to confirm this effect for the S/F systems with a good contact between metallic F and S layers made of ordinary metals and standard ferromagnets (see, e.g., recent reviews [1–3] and references therein). In spite of different values of the magnitude of the spin-valve effect $\Delta T_c = T_c^{\text{AP}} - T_c^{\text{P}}$ ($\Delta T_c = 10$ mK in Ref. [4], $\Delta T_c = 20$ mK in Ref. [5] and $\Delta T_c = 120$ mK in Ref. [6]), the full switching between the superconducting and normal states has been realized only in a few cases [7, 8] because ΔT_c was usually smaller than the width of the superconducting transition δT_c .

Very recently, Singh *et al.* reported [9] the observation of a colossal triplet spin-valve effect for the S/F1/N/F2 structure made of amorphous MoGe, Ni, Cu, and CrO_2 as the S, F1, N, and F2 layers, respectively. This structure demonstrated variation of T_c by ~ 1 K when changing the relative alignment of the two F layers. It was shown that the optimal operational field for this device is of the order of 20 kOe. Gu *et al.* [10, 11] reported $\Delta T_c \sim 400$ mK for Ho/Nb/Ho trilayers. Also in this case the parallel configuration of magnetizations was reached at a field of

~ 10 kOe. The high operational fields of these spin valves are disadvantageous for the superconducting spintronics. Besides, the physical reasons for large values of ΔT_c for spin valve based on half-metals are not yet theoretically explained. This calls for elaboration of classical spin-valve structures which use standard ferromagnets (Fe, Co, Ni) and their alloys with good electrical contacts between all layers and for which theoretical understanding of the operational principle is available.

Oh *et al.* [12] proposed theoretically a metallic F1/F2/S multilayer structure as a superconducting spin valve based on the S/F proximity effect. In our previous works on the $\text{CoO}_x/\text{Fe1}/\text{Cu}/\text{Fe2}/\text{In}$ structure we have demonstrated a full switching between the normal and superconducting states [7] and observed the sign-changing oscillating behavior of the magnitude of the spin valve effect ΔT_c on the thickness of the Fe2 layer [13, 14]. With that the F1/F2/S structure was experimentally established as a spin valve.

Recent theories (see, e.g., reviews [1–3, 15–18]) predict that at certain conditions a long-range triplet component (LRTC) in the superconducting condensate can arise in the S/F structure. The generation of the LRTC in the F1/F2/S spin-valve structure should manifest itself as a minimum of T_c at noncollinear configuration of magnetizations [19]. We have obtained experimental confirmation of this prediction by studying the $\text{CoO}_x/\text{Fe1}/\text{Cu}/\text{Fe2}/\text{Pb}$ spin valve structure [20]. The observed angular dependence of T_c was caused by a combination of the conventional and triplet components of the condensate. An indication of the triplet contribution to the magnitude of the superconducting spin-valve effect has been also observed in Refs. [4–6, 21, 22].

A crucial question of fundamental and application-related importance is whether it is possible to observe and even utilize an “isolated” triplet spin-valve effect. At first glance, it seems to be unrealistic, since LRTC arises entirely from the singlet component and cannot exist without it. However, here we experimentally demonstrate an “isolation” of the LRTC. We achieved it by exploiting the oscillatory behavior of $\Delta T_c(d_{F2})$ that effectively suppresses the conventional spin-valve effect by an appropriate choice of the F2-layer thickness d_{F2} . Furthermore, we succeeded to utilize the LTRC for the operation of the spin-valve and demonstrate the full switching effect for the superconducting current upon changing the mutual orientation of the magnetizations of F1 and F2 layers from AP to the *orthogonal* orientation. Finally, we have substantially improved the theoretical analysis by employing a fully quantitative approach for calculation of the T_c suppression due to the proximity effect.

To investigate the proximity-induced triplet pairing we measured the magnitude of the spin-valve effect in the dependence of the Py2-layer thickness for the $\text{CoO}_x/\text{Py1}/\text{Cu}/\text{Py2}/\text{Cu}/\text{Pb}$ multilayer grown on the MgO (001) substrate. Here Py denotes permalloy $\text{Ni}_{0.81}\text{Fe}_{0.19}$. The choice of Py for ferromagnetic layers appears crucial. As will be shown below, Py due to a smaller value of the exchange splitting of the conduction band in comparison with Fe shifts the maximum of ΔT_c towards a larger F-layer thickness and yields much larger ΔT_c for the spin-singlet and spin-triplet channels. In addition, the use of Py decreases the switching field by a factor of 6 in comparison with Fe as a magnetic layer. This enables to avoid any possible partial depinning of the bias Py1 layer by the switching field.

Optimization of the preparation conditions of the samples and their characterization are described in Ref. [23]. The optimal thickness of the Pb layer $d_{\text{Pb}} = 70$ nm was determined from the $T_c(d_{\text{Pb}})$ curve measured at a constant $d_{\text{Py1}} = 5$ nm, which is much larger than the penetration depth ξ_h of Cooper pairs into ferromagnetic Py. Basing on our data on $T_c(d_{\text{Py}})$ at fixed d_{Pb} we estimate this value as $\xi_h \simeq 1.1$ nm. At a large Pb layer thickness, T_c slowly decreases with decreasing d_{Pb} . Below $d_{\text{Pb}} \sim 120$ nm, T_c value starts to decrease rapidly. At $d_{\text{Pb}} \leq 40$ nm, T_c is less than 1.5 K. At small $d_{\text{Pb}} \leq 70$ nm the width of the superconducting transition curve gets extremely large, of the order of 0.4 K. Bearing in mind that the influence of the magnetic part of the structure gets stronger as the S-layer thickness approaches the superconducting coherence length ξ_s ($\xi_s \simeq 40$ nm for our samples), we have chosen $d_{\text{Pb}} = 70$ nm as a compromise value.

Earlier we revealed that the F1-layer thickness at a fixed d_{F2} does not significantly influence ΔT_c for the sample set CoO_x (2.5) / Fe1 (d_{Fe1}) / Cu (4) / Fe2 (d_{Fe2}) / Cu (1.2) / Pb (60 nm) and that a thin Cu (1.2-nm) interlayer between F2 and S layers is completely transparent

for the Cooper pairs [24].

All spin-valve structures were magnetically characterized using a standard 7-T VSM SQUID magnetometer (Fig. 1). First, the samples were cooled from 300 to 10 K in the presence of the in-plane magnetic field +4 kOe. At 10 K the magnetic field was varied from +4 kOe to -4 kOe and back again. During this variation, the in-plane magnetic moment of the sample was measured [Fig. 1(a)]. It turns out that for most of the $\text{CoO}_x/\text{Py1}/\text{Cu}/\text{Py2}/\text{Cu}/\text{Pb}$ samples the saturation field of the Py2 film is of the order of 200 Oe [Fig. 1(b)]. It can also be seen from this figure that the switching field $H_0 = \pm 150$ Oe is sufficient to sustain a homogenous magnetization for the Py2 layer following the switching field direction without formation of the domain structure [14]. At the same time the magnetization of the Py1 layer remains fixed up to the operating field of the order of -2.5 kOe due to its pinning by the antiferromagnetic CoO_x layer (Néel temperature $T_N \sim 250$ K). This result is very similar to those observed for $\text{CoO}_x/\text{Fe1}/\text{Cu}/\text{Fe2}/\text{Cu}/\text{Pb}$ structure (see Fig. 4 of Ref. [24]).

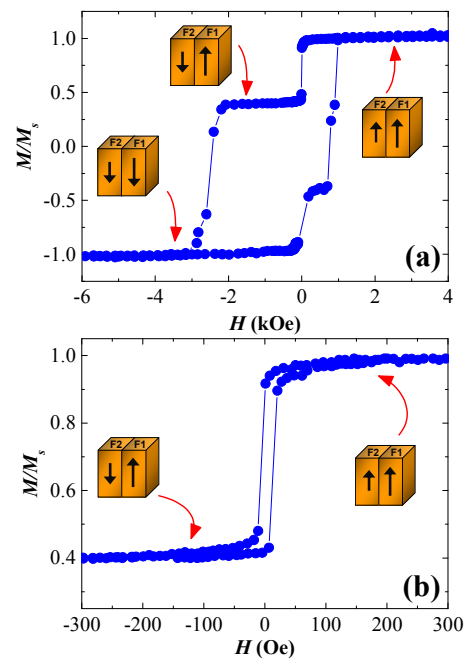


FIG. 1. (Color online). (a) Magnetic hysteresis loop for the CoO_x (3) / Py (3) / Cu (4) / Py (1.2) / Cu (2) / Pb (70 nm) structure. (b) The central part of the minor hysteresis loop for this sample due to the reversal of the magnetization of the free Py2 layer. Arrows denote mutual orientation of the magnetization of the two Py layers.

For the transport study we used another system which enables very accurate control of the magnetic field acting on the sample. We have combined the electrical setup with a vector magnet that enables a continuous rotation of the magnetic field in the plane of the sample. To avoid the occurrence of the unwanted out-of-plane component

of the external field, the sample plane position was always adjusted with an accuracy better than 3° relative to the direction of the dc external field. By preserving the in-plane orientation of the external field we avoid any noticeable angular dependent change in the demagnetization field. The magnetic field value was measured with an accuracy of ± 0.3 Oe using a Hall probe.

For the set of spin-valve samples with various d_{Py2} , we studied the dependence of the superconducting transition temperature T_c on the angle α between the direction of the cooling field and the external magnetic field both applied in plane of the sample. For the CoO_x (3) / Py (3) / Cu (4) / Py (0.6) / Cu (2) / Pb (70 nm) structure we observed a large magnitude of a conventional spin valve effect with $\Delta T_c = 110$ mK [Fig. 2(a)].

One can see in Fig. 2(a) that upon changing the mutual orientation of magnetizations by a gradual in-plane rotation of the magnetic field from the P ($\alpha = 0^\circ$) to the AP ($\alpha = 180^\circ$) state, the T_c does not change monotonically but passes through a minimum. According to theory, a characteristic minimum in the $T_c(\alpha)$ dependence (which is most pronounced if it takes place near $\alpha = 90^\circ$ but may not be necessarily located exactly at this angle) is a fingerprint of the LRTC [19]. Though the triplet component is inherent in the case of noncollinear magnetizations, assuming for a moment its absence, one would expect the $T_c(\alpha)$ dependence to be monotonic. From general symmetry, $T_c(\alpha)$ must behave as α^2 and $(\pi - \alpha)^2$ when α deviates from 0 and π , respectively (since deviations to both sides of this values are physically equivalent, and we expect $T_c(\alpha)$ to be an analytic function). One would then arrive at a simple angle-dependent superposition of the limiting values of T_c^P and T_c^{AP} , which reads as $T_c^{(\text{ref})}(\alpha) = T_c^P \cos^2(\alpha/2) + T_c^{AP} \sin^2(\alpha/2)$. This dependence is shown by the dashed line in Fig. 2(a) and we consider it as a reference curve. Deviation of the actual T_c from the reference curve, shown in Fig. 2(b), demonstrates the LRTC contribution to the ΔT_c . We denote it as ΔT_c^{trip} . Figure 2(a) shows that the difference in T_c between AP and perpendicular orientations of magnetizations amounts to as much as 130 mK. It means that the LRTC significantly contributes to the spin-valve effect.

In our previous works [13, 14, 20, 24], we compared our experimental data for T_c with the effective boundary parameter W that enters the theory [19] and determines how strongly the F part of the system suppresses the superconductivity in the S layer. This approach allowed us to demonstrate a good qualitative agreement between theory (W) and experiment (T_c) without actually calculating the critical temperature. Here, we apply the method of Ref. [19] (extended to the case of arbitrary S/F interface transparency [25]) for direct fitting of our data on $T_c(\alpha)$, as shown in Fig. 2(a) by a solid line. Here we used the following set of parameters: the superconducting coherence lengths for the S and the F layers $\xi_s = 41$ nm and $\xi_f = 13$ nm; the S layer thick-

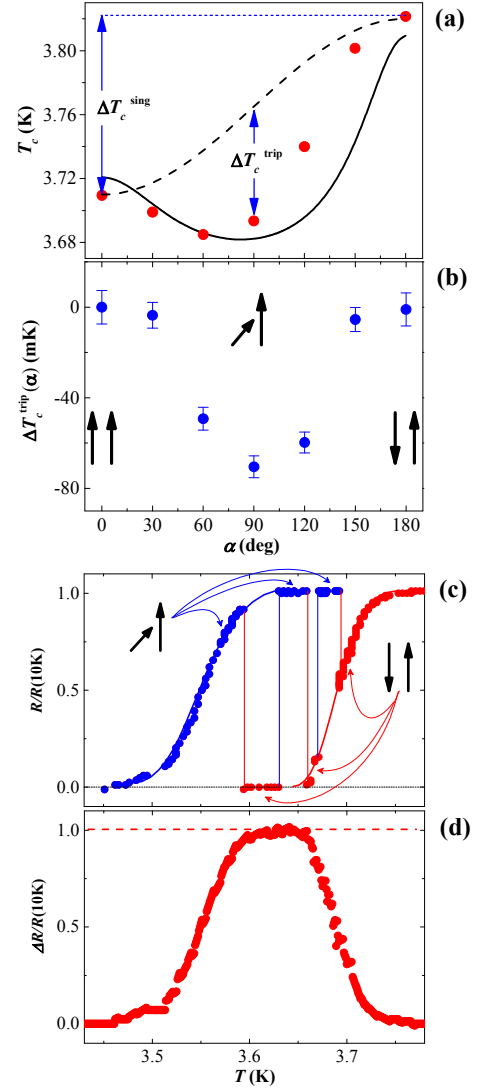


FIG. 2. (Color online). Spin-valve effect for the CoO_x (3) / Py (3) / Cu (4) / Py(0.6) / Cu (2) / Pb (70 nm) structure. (a) Angular dependence of T_c measured at a field $H_0 = 150$ Oe (circles), estimated α dependence of the singlet spin valve effect ΔT_c^{sing} (dashed line), and the theoretical result according to Ref. [19] (solid line) (see the text). (b) Difference ΔT_c^{trip} between the actual T_c and the reference curve [dashed line in panel (a)]. (c) Switching between normal and superconducting states in the CoO_x (2.5) / Py (3) / Cu (4) / Py (1.0) / Cu (2) / Pb (70) spin valve sample by sweeping slowly the temperature within ΔT_c and changing the direction of magnetic field α between 180° (closed circles) and 90° (opened circles). (d) Temperature dependence of $\Delta R = R(\alpha = 90^\circ) - R(\alpha = 180^\circ)$ demonstrating the full switching in the temperature range $3.6 \div 3.66$ K.

ness $d_s = 73.5$ nm; the bulk critical temperature of the S layer $T_{cs} = 7.2$ K; the transparency parameters $\gamma = 0.734$ and $\gamma_b = 1.8$; and the exchange field acting on the spins of conduction electrons in the F layer $h = 0.3$ eV. We found that the theoretical description requires much smaller $d_{Py2} = 0.3$ nm than the nominal $d_{Py2} = 0.6$ nm.

There are several possible reasons for that. First, the Py2 layer is sandwiched by two Cu layers. Due to the interdiffusion the effective thickness may be reduced down to 0.3 nm. Second, the theory does not take into account details of the band structure of the materials constituting our structure. It can also be that the dirty-limit conditions assumed by the theory, are not fully satisfied in our system. In any case, as seen in Fig. 2(a), theory fits experimental data satisfactorily at reasonable values of parameters.

The magnitude of the spin-valve effect upon changing the mutual orientation of magnetizations from AP to the orthogonal one exceeds the width of the superconducting transition curve. Therefore it is possible to switch off and on the superconducting current flowing through our samples *completely*, as demonstrated in Fig. 2(c). A complete on/off switching of the resistance of the sample due to the combination of the triplet spin valve effect and ordinary spin valve effect is shown in Fig. 2(d).

From the data in Fig. 2 it is obvious that the LRTC aids in reaching larger spin-valve effect but still the triplet effect interferes with the conventional one. As we have previously shown, the amplitude of the conventional spin-valve effect can be suppressed to zero for certain thickness d_{F2} due to the oscillating behavior of $\Delta T_c(d_{F2})$ [13, 14]. These oscillations are caused by the interference in the F2 layer of the pair condensate wave function coming from the F2/S interface with the one reflected from the F1/F2 interface. For $\text{CoO}_x/\text{Fe1}/\text{Cu}/\text{Fe2}/\text{Cu}/\text{Pb}$ structure ΔT_c reduces to zero for $d_{Fe2} \simeq 0.8 \div 1$ nm [24, 26]. From the analysis of our present data on the $T_c(d_{Py})$ dependence we conclude that the penetration depth of the Cooper pairs into Py exceeds the value obtained for Fe by 30%. Therefore $\Delta T_c = 0$ in the samples with Py should be reached at $d_{Py2} \sim 1 \div 1.7$ nm. Indeed, such a sample with $d_{Py} = 1.7$ nm prepared in the present work demonstrates the “isolated” triplet spin-valve effect (Fig. 3). Obviously, the conventional singlet spin-valve effect van-

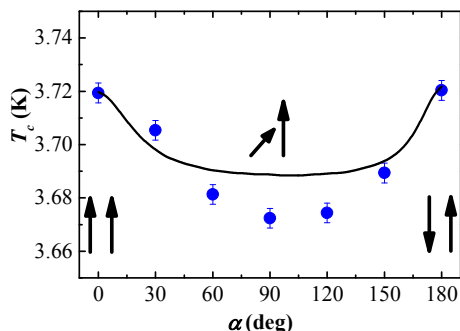


FIG. 3. (Color online). Sample CoO_x (3) / Py (3) / Cu (4) / Py (1.7) / Cu (2) / Pb (70) with the zero conventional spin-valve effect. Angular dependence of T_c caused by the LRTC is shown by circles, and the solid line is the theoretical curve according to Ref. [19] (see the text).

ishes, $\Delta T_c = T_c^{\text{AP}} - T_c^{\text{P}} = 0$. At the same time at non-collinear orientation of magnetizations of the Py1 and Py2 layers the $T_c(\alpha)$ dependence exhibits a minimum. In accordance with theory [19] for this sample the amplitude of ΔT_c^{trip} is smaller than for the sample in Fig. 2 because of the larger value of d_{Py2} . That is because the LRTC is generated when Cooper pairs reach the Py1 layer after leaving the superconductor and penetrating into the Py2 layer. As a result, the thicker Py2 layer suppresses both the singlet component and the LRTC that is generated from the singlet one. Fitting of these data was performed using the parameters of theory which we already used in calculation of the ΔT_c dependence in Fig. 2(a). The only difference was in d_{Py2} , which was taken 1.7 nm.

The possibility to isolate the spin-triplet component which we have shown in this work is remarkable. The targeted engineering of S/F heterostructures where peculiarities of the interference of the superconducting pairing wave function make the spin-singlet component ineffective from the viewpoint of influencing T_c appears to be straightforward. Such heterostructures are promising candidates for building spintronic devices where the functionality of the triplet Cooper pairs carrying not only charge but also the spin polarization over a long distance is essential. Generation, control, and manipulation of such spin supercurrents in S/F multilayers appear thus as an emerging field of undisputable importance both for fundamental physics and for material research.

In conclusion, we have experimentally investigated the long-range proximity-induced triplet superconductivity in the F1/F2/S structure. Our main experimental result is the “isolation” of the triplet spin-valve effect by exploiting the oscillating behavior of the ordinary spin-valve effect. Furthermore, we have shown that the spin-triplet component can be utilized for the operation of the spin valve and demonstrate the spin-triplet-assisted full switching effect for the superconducting current. On the theory side, we have successfully applied a fully quantitative approach for calculating the suppression of T_c due to the proximity effect in the studied heterostructures.

This work was supported by the Deutsche Forschungsgemeinschaft through Grant LE 3270/1-1. It was also partially supported by RFBR (Grants No. 13-02-01389-a and No. 14-02-00350-a), by programs of the RAS, by the Ministry of Education and Science (Russian Federation), and by the program “5top100.”

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